ULTRASONIC CHARACTERIZATIONS OF SOLIDS HOLDUP IN A BUBBLE COLUMN REACTOR

Yee Soong*, Daniel J. Fauth and James P. Knoer

U. S. Department of Energy, National Energy Technology Laboratory,

P.O. Box 10940, Pittsburgh, PA 15236, U.S.A.

Tel. : (412) 386-4925

Fax : (412) 386-4810

E-mail : Soong@netl.doe.gov

Prepared for Chemical & Engineering Technology

* To whom all correspondence should be addressed

1 Introduction

For the optimum design and operation of gas-liquid-solid three-phase reactors, the degree of dispersion of the solid (catalyst) in the reactor must be measured, understood, and controlled. Various methods to measure this parameter, including optical, direct sampling and the static pressure method, have already been developed and used experimentally. However, these methods have inherent disadvantages. Accurate means of diagnosing the solids holdup are needed. The ultrasonic technique offers more practical applications in that it is noninvasive, nondestructive, nonhazardous, rapid, and potentially applicable to high-temperature flow in high-pressure, opaque-wall reactors. The utilization of ultrasonic techniques for slurry characterizations has received considerable attention recently [1-5]. Some ultrasonic techniques are based on the principle of scattered acoustic pulses [1-4]. The other uses of the ultrasonic Doppler technique to characterize the local bubble rise velocity in a bubble column reactor [5]. Recently, a method involving the measurement of ultrasound transmission has been reported in a slurry-phase stirred-tank reactor which offers the possibility of using the ultrasonic technique to measure solids holdup in a three-phase slurry reactor [6,7]. The ultrasonic transmission uses measurements of the velocity and attenuation of the sound wave which travels directly through the slurry sample. When the velocity of sound in a liquid is significantly different from that in a solid, a time shift (a velocity change) in the sound wave can be detected when solid particles are present relative to that for the pure liquid. Fig. 1 shows how the detected sound wave is varied in time and in amplitude when solids are suspended in the liquid. The arbitrary first distinct zero crossing time in liquid and in solid-liquid

are defined as t_l and t_x, respectively. The travel time between the transmitter and receiver in the liquid is defined as t_o. Okamura et al., [6] and Soong et al., [7] used a continuous stirred-tank reactor to correlate the solids holdup to the relative time shift $((t_l-t_x)/t_o)$. The arbitrary first distinct zero crossing time, t_x, can also be determined in gas-liquid and gas-liquid-solid systems. Those researchers [6,7] also indicated that the t_x in the gas-liquid-solid system is not affected by the presence of gas bubbles and is fixed at the same position as in the solid-liquid system. Furthermore, the application of the measurement of ultrasound transmission for gas holdup [8-10] and for gas holdup as well as low concentration of solid (up to 3 wt.%) under limited superficial gas velocities (up to 3 cm/s) in a slurry-bubble-column reactor has been reported [11,12]. This leads to the study of using the ultrasonic technique for the measurement of solids holdup in a three-phase bubble column reactor over a wide range of superficial gas velocities and solids holdup.

2 Experimental

The detailed information of the bubble-column-reactor in which the ultrasonic investigation was conducted has been reported elsewhere [10,13]. The transparent acrylic bubble-column-reactor has an internal diameter of 8.89 cm and a height of 290 cm (Fig. 2). The ultrasonic signals are transmitted at 33 cm above the bottom of the gas distributor, which is a perforated-plate gas distributor with 15 x 1-mm diameter holes. Experiments were conducted in batch-mode operation (stationary liquid - water and continuous flow of gas - nitrogen). The nitrogen flow was controlled electronically to a maximum of 12 cm/s through a mass-flow controller. Glass beads from Cataphote, Inc., (10-37 μm in diameter with density of 2.46 g/cm^3) were used as the solid in the slurry. The solids holdup (solid weight/total slurry weight) was varied from 5 to 30 wt.% for each

nitrogen flow in the reactor. To evaluate the accuracy of the ultrasonic technique for solids holdup measurement, an independent slurry sampling device was installed. The measurement was conducted by inserting a stainless steel tubing (0.775 cm. I.D.) horizontally into the center of the column at 0.635 cm above the path of the ultrasonic transmission. The ultrasonic transmitter/receiver and the solid sampling device are positioned such that both means are measuring approximately the same hydrodynamic phenomena as shown enlarged areas in Fig. 2. Both the transmitter and receiver were mounted directly inside the reactor wall at 33 cm above the gas distributor.

3 Results and Discussion

Fig. 3 illustrates the effects of solids holdup on the transit time measured at 33 cm above the gas distributor in the glass beads/nitrogen/water system at different superficial gas velocities (SGVs). In this experiment, the SGV was systematically varied at any given initial solids holdup of 5, 10, 20, and 30 wt.% in the bubble column reactor. In general, the transit time varies with the variation of the superficial gas velocity for the SGV of 4 cm/sec or less at any given initial constant solids holdup loading in the reactor. The transit time was relative constant when the SGV is 4 cm/sec or higher. The transit times are around 71.96, 71.6, 71.12 and 70.88 μs for the solids holdup of 5, 10, 20, and 30 wt.% respectively, when the SGV is 4 cm/sec or higher. Therefore, the transit time can be utilized to determine the solids holdup when the column is operated in a complete suspension mode. The fluctuation of the transit time when the SGVs is 4 cm/sec or less may attribute to the both partial sedimentation and other factors which are under investigation. The fractional change of transit time ( ) $t/t_o = (t_r - t_x)/t_o$ can be calculated on each individual transit

time in Fig. 3. From the fractional change of transit time, the solids holdup can be determined from the previous calibrated curve obtained from a stirred tank reactor (Fig. 11. in Soong et. al.[7]). The determined solids holdup from these procedures and the solids holdup determined by the direct sampling are illustrated in Fig. 4. The solids holdup clearly varies with the SGVs. For a 30 wt.% solid loading in the bubble column, the solids holdup varies from 0.466 to approximately 0.393 as the SGV increased from 0.53 to 2.68 cm/sec. The occurrence of the partial semination in the column was also observed in the transparent acrylic column under this condition. At 2.685 cm/sec gas velocity, the solids holdup determined by the ultrasonic technique is 0.393 compared to 0.323 by the direct sampling technique. At elevated SGV of 3.22 cm/sec or higher, the solids holdup is approximately 0.369 as determined by the ultrasonic technique compared to 0.337 determined by the direct sampling technique. For 20 wt.% solid loading in the bubble column, when the SGV increases to 3.22 cm/sec or higher, the ultrasonic technique estimates the solids holdup at 0.295. However, the solids holdup determined by the direct sampling technique is between 0.2044 and 0.246 under these conditions. For 10 wt.% solid loading in the bubble column, the holdup profile is similar to that of 20 wt.%. At the SGV of 1.61 cm/sec, the solids holdup determined by the ultrasonic technique is 0.092 compared with that of 0.102 determined by direct sampling. For a 5 wt.% loading conditions in the bubble column, when the SGV is 3.22 cm/sec or higher, the solids holdup is approximately 0.044 as determined by the ultrasonic technique. Furthermore, the solids holdup determined by direct sampling is between 0.0539 and 0.0286 under the same experimental conditions. The solids holdup measurements by the ultrasonic technique compared reasonably well with results obtained by the direct sampling techniques. Some

discrepancies observed between these two techniques are probably due to the nature of these techniques. The ultrasonic technique measures the average solids holdup in the ultrasound path while direct sampling determines the collected local solids holdup. The simple time-averaged method utilized in this study does not account for the sound refraction and reflection of the solid phase on the transit time[3]. Uchida et al. [11] and Warsito et al. [12] have reported some experimental results obtained from low concentrations of solids (up to 3 wt.%) and low gas velocity (up to 3 cm/sec). It is difficult to compare our data collected under high gas velocities and high solids holdup with those collected under different conditions. Warsito et al. [12] also proposed a theoretical model that related the solids holdup with the transit time ratio. This approach can be applied to further theoretical study.

4 Conclusion

An ultrasonic transmission technique has been developed to measure solids holdup in a gas-liquid-solid bubble column reactor. The results presented in this study show that the transit time of an ultrasonic signal is influenced by the variation of solids holdup and the operating conditions in the bubble column. The transit time can be correlated to the solids holdup. The ultrasonic technique is potentially applicable to high-temperature, non-transparent fluids in high-pressure, metallic reactors and, with some modifications for solids holdup measurements, are applicable in slurry-bubble-column reactors.

Acknowledgments

The authors express their appreciation to R. R. Anderson for his assistance.

Symbols used

t_l Arbitrary first distinct zero crossing time in liquid, s

t_X Arbitrary first distinct zero crossing time in gas-liquid, solid-liquid or gas-liquid-solid system, s

t_O Arbitrary travel time of the sound wave between the transmitter and the receiver in liquid, s

References

1. US Pat., 4, 580, 444, 1986, Micro PureSystems, Inc., (Inv.: Abts, L. R.; Dahi, P. H.).

2. Behrman, C. E.; Larson, J. W. On-line Ultrasonic Particle Monitoring of Brewing Operations. *MBAA Tech. Quart.* 24 (1987) pp. 72-76.

3. Bonnet, J. C.; Tavlarides, L. L. Ultrasonic Technique for Dispersed-Phase Holdup Measurements, *Ind. Eng. Chem. Res.* 26 *(*1987) pp. 811-815.

4. US Pat., 4, 527, 420, 1985, Micro Pure System, Inc., (Inv.: Foote, K. G).

5. Hilgert, W.; Hofmann, H. Characterization of Gas Phase Flow in Bubble Columns at Low Superficial Gas Velocities with the Aid of Ultrasonic Doppler Technique. *Ger. Chem. Eng. 9* (1986) pp. 180-190.

6. Okamura, S.; Uchida, S.; Katsumata, T.; Iida, K. Measurement of Solids Holdup in a Three-Phase Fluidized Bed by an Ultrasonic Technique. *Chem. Eng. Sci*., 44 (1989) pp. 196-198.

7. Soong, Y.; Blackwell, A. G.; Schehl, R. R.; Zarochak, M. F.; Rayne, J. A. Ultrasonic Characterization of Three-Phase Slurries, *Chem. Eng. Com.*, 138 (1995) pp. 213-224.

8. Chang, J. S.; Ichikawa, Y.; Irons, G. A.; Morala, E. C.; Wan, P. T. Void Fraction Measurement by an Ultrasonic Transmission Technique in Bubbly Gas-Liquid Two-Phase Flow. Measuring Techniques in Gas Liquid Two-Phase Flows, Delhaye, J.M. and Cognet, G., Eds, Springer-Verlag: New York, (1984) pp.319-335.

9. Bensler, H. P.; Delhaye, J. M.; Favreau, C. Measurement of Interfacial Area in Bubbly Flows by Means of an Ultrasonic Technique. *Proc. ANS Natl. Heat Transfer Conf.* , 1987, pp. 240-246.

10. Soong, Y.; I. K. Gamwo.; A. B. Blackwell.; Harke, F. W.; Schehl, R. R.; Zarochak, M. F. Ultrasonic Characterizations of Gas Holdup in a Bubble Column Reactor, *Chem. Eng. Comm*.158 (1997) pp. 181-192.

11. Uchida, S.; Okamura, S.; Katsumata, T. Measurement of Longitudinal Distribution of Solids Holdup in a Three-Phase Fluidized Bed by Ultrasonic Technique, *Can. J. of Chem. Eng.* 67 (1989) pp.166-178.

12. Warsito, A.; Maezawa, A.; Uchida, S.; Okamura, S. A Model for Simultaneous Measurement of Gas and Solids Holdup in a Bubble Column Using Ultrasonic Method. *Can. J. of Chem. Eng*. 73 (1995) pp.734-743.

13. Soong, Y., Gamwo, A. G., Harke, F. W., and Ladner, E. P., Ultrasonic Characterization of Slurries in a Bubble Column Reactor, *Ind. Eng. Chem. Res.* 38 *(*1999) pp .2137-2143.

LIST OF FIGURES